Catalysts for Fuel Cell Transportation and Hydrogen Related Uses

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The global energy and transportation landscapes are changing rapidly, and that brings with it evolving opportunities and catalyst research needs for hydrogen and fuel cells.

Polymer electrolyte fuel cells often have been promoted as one—if not the—solution for clean, sustainable transportation. While a review of the history of fuel cell transportation development over the past few decades reveals a tumultuous development trajectory (including major commercial and industrial investments of tens of billions of dollars; overselling of how soon and in what quantity and cost fuel cell vehicles would be available; and strong detractors from the technology and approach), the technology has advanced significantly and reflection on recent trends reveals rapid progression. For light-duty transportation, the past few years have seen manufacturing volumes increase to several thousands of vehicles per year as a hydrogen refueling infrastructure is rolled out in parallel in select global locations (Germany, Japan, California)[1]. These efforts have demonstrated the technological viability of fuel cell vehicles and allowed for announced expansions from the 10s to 100s of thousands units/yr production level (Toyota revealed plans to go to 30,000 units/yr in 2020[2], and Hyundai announced a $6.7 billion investment in the technology targeting 40,000 units/yr in 2022 and 700,000 units/yr in 2030[3]). These major investments are reflections of the technological advances that have occurred at the vehicle level, the developing infrastructure that allows for vehicle fueling, and the evolving societal and business concerns for zero-emission/electrified transportation.

A major enabling aspect of commercially relevant light-duty fuel cell vehicles has been the dramatic reduction of Pt loading in anode and cathode electrocatalysts while also improving cell performance and durability. The U.S. Department of Energy supports analyses to baseline and track the cost of fuel cell systems and catalyst loadings. Figure 1 presents a summary of these studies dating back

to 2002 for 500,000 units/yr production volume[4]. By plotting fuel cell system costs and catalyst loadings from 2002 to 2017 together, it is immediately apparent there has been a dramatic reduction in both over time (an 84% reduction in system cost and 88% reduction in catalyst loading), and the trends parallel each other reasonably well. The observed change in system cost reflects not just catalyst cost improvements but also cost gains in non-catalyst areas. While the decrease in catalyst loading is substantial, it is even more impressive considering that cell power density doubled and durability increased substantially. Improving the cost, performance and durability together has been critical in enabling efficient, effective Pt use.

In 2002, Pt loading was projected to be 1.0 mg/cm^2 active area (about 80 g per vehicle); in 2017 this projection decreased to 0.125 mg/cm^2 (about 10 g per vehicle). Assuming \$30/g Pt prices (near historical averages), 10 g of Pt would add about \$300/vehicle, an important cost driver but not an overwhelming percentage of vehicle or fuel cell system cost. Furthermore, at 10 g per vehicle the platinum group metal (PGM) content of the fuel cell stack approaches parity with advanced catalytic converter PGM loadings for internal combustion engine vehicles (as fuel cell vehicles are zero-emission vehicles they have no need for further catalytic conversion). Current projections for fuel cell system cost are near \$45/kW at large volume with proposed targets of \$30/kW to reach full parity with internal combustion vehicles, and Pt remains a strong cost component, so further advances are still important. However, the significant advances to date have dramatically improved commercial viability.

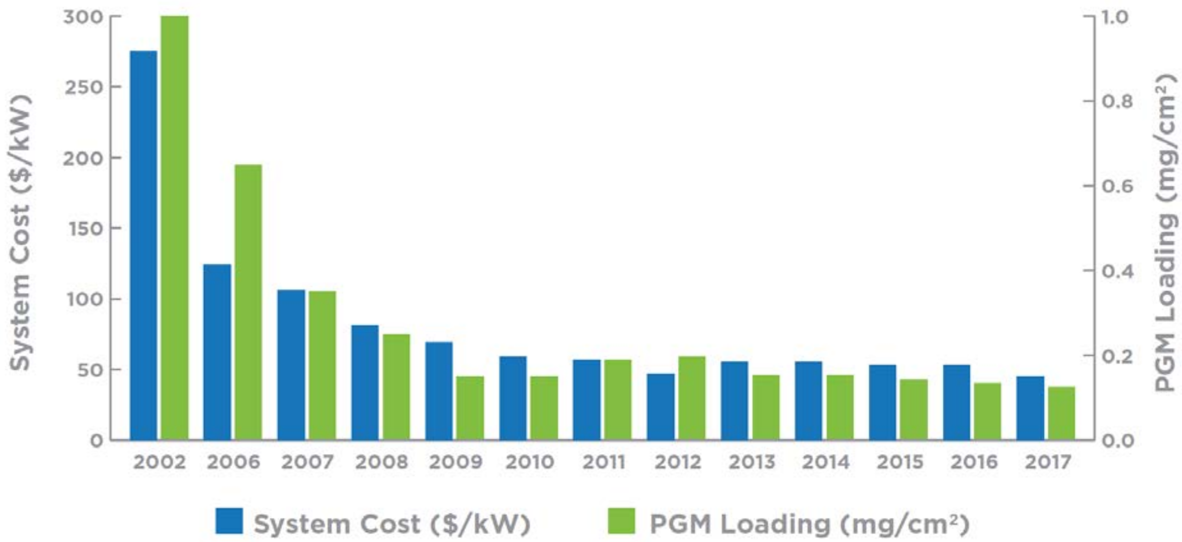

Fig 1. System cost and catalyst loading. Modeled fuel cell system costs and catalyst Pt group metal (PGM) loading for light-duty vehicle polymer electrolyte fuel cell systems manufactured at 500,000 units per year[4] have shown dramatic decreases from 2002 levels. The observed decreases reasonably parallel one another.

Hydrogen beyond transportation

While technological advances have improved the commercial viability of fuel cell vehicles, other factors also are influencing the potential role of hydrogen and fuel cells in our energy system. The societal megatrends that are impacting, or have the potential to impact, hydrogen and fuel cells include: increasing global population; increasing recognition and regulation surrounding emissions; decreasing costs of variable renewable energy; increasing urbanization; and changing societal views on transportation and autonomous vehicles. These factors, at least in part, have led to multiple efforts and programs that pursue hydrogen as a key element of our evolving energy system.

In the United States, H2@Scale is a U.S. Department of Energy effort focused on increasing hydrogen utilization for energy-system-wide benefits[5]. An illustrative H2@Scale energy system is presented for visualization purposes in Figure 2. This schematic highlights the ability of hydrogen to

serve as a clean energy intermediate, linking power generation (fossil, nuclear, renewable) to other energy sectors (industry and transportation). In this way hydrogen helps to balance mismatch in power generation and demand while allowing for cross-sectoral and cross-temporal impact. Other related visions and programs exist, for example, Power-to-X in the EU,[6] the Hydrogen Society in Japan[7] and the Hydrogen Council's Hydrogen: Scaling Up roadmap[8]. All these efforts share the common link that hydrogen is a critical, scalable intermediate that can address many of the current energy system's limitations. A primary argument against fuel cell vehicles has been the cost and availability of the hydrogen infrastructure to produce, store and move hydrogen cost effectively. As the value and need for hydrogen to service other areas beyond transportation is better understood, it increases the potential impact in transportation.

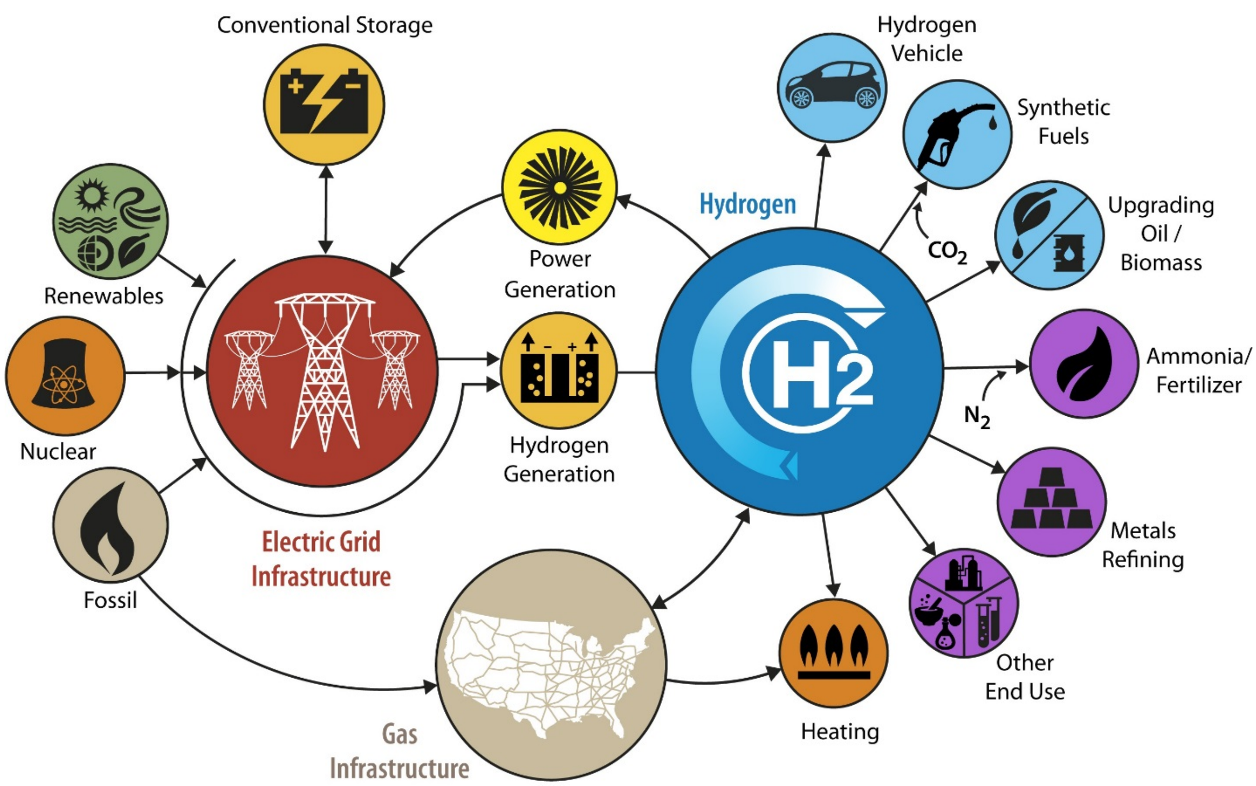

Fig 2. Illustrative H2@Scale energy system. The schematic depicts hydrogen's potential parallel role in the energy system as a clean, efficient energy carrier like the electrical grid and natural gas system.

Notably, hydrogen provides benefits in linking energy generation with end use and in the ability to shift energy across sectors, time and distance.

While this Comment focuses largely on fuel cell electrocatalysts for transportation, there are multiple areas where catalysis and electrocatalysis will be important in an H2@Scale energy system. Hydrogen generation is particularly dependent on catalyst improvements. Improvements in electrolysis catalyst cost, performance and durability are a major R&D need. Over the past 20 years, 10s of billions of dollars has been invested to develop the systems and advance the science of fuel cells, while investment in electrolysis has been orders of magnitude lower. The attributes of fuel cell systems—relatively high efficiency, ability to respond to dynamic operation including start/stop, and projected lifetimes of several years—project well onto (low-temperature) electrolysis systems. There are some key differences between fuel cells and electrolysis systems that require specific focus, including mass transport concerns (water consumption and gas generation for electrolyzers vs. gas consumption and water generation for fuel cells), higher (differential) pressure operation, and higher-voltage operation (which significantly impact materials choices). Many of the approaches to advancing fuel cell science, and the scientific community that has played a major role in these advancements, are likely to be able to positively impact electrolysis R&D. Beyond low-temperature electrolysis, high-temperature electrolysis and catalytic routes that yield hydrogen from methane or other fossil fuels while perhaps producing other carbonaceous value-added products are also of high interest.

Beyond hydrogen generation, there are significant catalysis challenges on how to best use and/or transform hydrogen once it is produced. While hydrogen is already used in large quantities today for applications like fuel upgrading and ammonia production, processes like Fischer–Tropsch, Sabatier, and Haber–Bosch have largely been unchanged over several decades. They suffer from efficiency and integration concerns and it is challenging to efficiently and cost-effectively scale them down in size.

These and related processes will continue to gain importance as the energy system continues to evolve from one almost solely based on fossil fuels to one with more renewable and/or nuclear generation.

Evolving transportation sector

Within the transportation sector several major changes are taking place. These include a focus on zero-emission vehicles to address local air quality issues and the increased efficiency of electrified propulsion. Societal driving habits are also evolving with ride sharing apps, public transportation, and the development of autonomous vehicles offering the potential for disruptive changes as transportation may be viewed more as a service. Over the past few decades personal sentiment toward transportation and vehicle ownership has changed significantly, and these changes are likely to continue and possibly accelerate.

One significant impact to the potential market for fuel cell vehicles has been the impressive progress and improvement of batteries and battery vehicles. The cost of battery vehicles has dropped significantly, and the performance of batteries has improved. From zero-emission vehicle and electrified drivetrain standpoints, battery and fuel cell vehicles both offer similar benefits, but each type has different attributes that make it better at potentially filling specific market roles. Battery vehicles have existing infrastructure for charging, an established manufacturing base with larger demonstrated capacity, and efficient charging and propulsion. Hydrogen-fueled fuel cell vehicles can refuel quickly and can decouple power from energy, resulting in benefits for heavy-duty and long-range transportation.

Fuel cell activity and interest in the heavy-duty transportation sector recently has increased significantly as the specific attributes of high energy density and fast refueling have been recognized. Much of the focus is on freight applications including long-haul and around ports. A few specific examples include Toyota announcing its Project Portal (employing light-duty fuel cell systems into heavy duty applications)[9], Nikola establishing contracts and a business plan to provide heavy-duty hydrogen-based transport across the United States[10], and efforts to establish hydrogen infrastructure for drayage

fleets at ports[11]. Beyond trucking, fuel cells are also being investigated for rail, marine and even air transportation applications. These potential transportation markets add to the markets that fuel cells currently serve (units deployed for materials handling/forklifts and stationary power or combined heat and power applications far outnumber fuel cell vehicles today)[12]. They also are important for the catalysis research community because the economic drivers are different for various applications, and the relative importance of performance and durability can be very different. For example, most research to date has focused on light-duty transportation, in which cost is the primary driver and small, nanoparticle Pt-alloy catalysts have sufficient durability with high performance. Applications such as heavy-duty transportation have longer life cycles and can tolerate higher capital cost. These different application needs will likely lead to additional R&D opportunities where factors like durability become weighted more heavily than cost.

Ongoing and future R&D needs

When discussing the development of fuel cell electrocatalysts, the focus is almost always on the cathode and the oxygen reduction reaction because the hydrogen oxidation reaction is exceptionally facile in acidic media on Pt[13]. Over the past few decades the development of advanced Pt-based catalysts has focused on several different approaches including alloys, size and shape control, nanostructured catalysts/facet control, extended surface catalysts, and skin composition/core-shell catalysts[14]. These approaches have shown different degrees of performance and durability improvements and many continue as active research areas.

PGM-free catalysts are a research area of particularly high interest, with significant effort globally to avoid PGM. However, the dramatic decrease in PGM content the community has demonstrated results in diminishing returns in achieving completely PGM-free status, and PGM-free catalysts will only be a replacement for PGM if performance and durability can be brought to the level of PGM catalysts for most applications. This is a significant challenge for carbon-based PGM-free catalysts

that has yet to be demonstrated, in particular for durability. However, these materials may hold promise as an active carbon replacement for support materials in PGM-containing electrodes and remain an ongoing area of research interest[15].

The performance of alkaline membrane fuel cells has improved tremendously in recent years, demonstrating significant improvements in power density and durability[16]. The promise of alkaline systems has always been their potential ability to decrease or completely avoid PGM catalysis. While reasonable performance has been shown for the oxygen reduction reaction, the overpotentials associated with hydrogen oxidation and the ability to limit PGM content at the anode remain research challenges.

The advances in fuel cell performance and durability are not solely due to the development of advanced catalysts. In fabrication of high-performance electrodes, issues beyond inherent catalytic activity including ion, electron and mass (hydrogen, oxygen and water) transport are critical. While electrocatalyst development has shown continual incremental progress, a clear breakthrough in fuel cell electrode R&D was the development and demonstration of the thin film electrode[17], achieved by incorporating polymer electrolyte into the catalyst-containing electrode, resulting in greatly improved ion, electron and mass transport characteristics. The thin film electrode approach has been largely unchanged despite the recognized impacts of catalyst-ionomer interactions and the mass transport limitations that fuel cells experience at high current and low loading[18]. Designing and architecting electrodes specifically for improved performance is an area that offers significant promise, as the highest catalytic activities demonstrated in ex situ characterization have fallen far short of the activities achieved in operating cells.

The view of hydrogen and fuel cell technology is changing rapidly as governments and industry better understand and articulate its value proposition. This is marking a step change for hydrogen and fuel cells from a technology of the future to the technology of today. Electrocatalyst improvements have

been critical in enabling advances in hydrogen and fuel cell technology and will continue to be a critical R&D area as the technology moves beyond acidic, polymer electrolyte systems for light-duty transportation to include efforts in other components (alkaline membranes, electrode fabrication) and in additional markets including heavy-duty transportation, hydrogen production, and end-use applications.

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